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**AUTHOR(S):** E.H. Essington  
E.B. Fowler  
W.L. Polzer

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RETENTION OF LOW-LEVEL RADIOACTIVE  
WASTE MATERIAL BY SOIL\*

by

E. H. Essington, E. B. Fowler, and W. L. Polzer

University of California  
Los Alamos Scientific Laboratory  
Los Alamos, NM

Abstract

Low-level radioactive wastes produced by users of radionuclides are generally disposed by shallow land burial. Reliance for containment is placed on characteristics of shallow geologic formations or soils; thus, effective waste management requires a knowledge of radioactive waste/soil interactions.

Because of the wide variations in soil and waste characteristics, the degree of radionuclide retention would be expected to vary; knowledge of that variation may be of value in predicting radionuclide mobility. This report discusses results of investigations of radioactive waste/soil interactions as they relate to radionuclide retention and its variability among soils and radionuclides.

In soil column leaching studies, radioactive waste solutions were applied to four different soil types;  $^{241}\text{Am}$ ,  $^{88}\text{Y}$ , and  $^{172}\text{Hf}$  were retained in the top four cm of soil with better than 90% retained by a protective surface sand layer. Less than 50% of the  $^{85}\text{Sr}$ ,  $^{137}\text{Cs}$ , and  $^{83}\text{Rb}$  was retained by the surface sand. No  $^{88}\text{Y}$ ,  $^{172}\text{Hf}$ ,  $^{85}\text{Sr}$ ,  $^{137}\text{Cs}$ , or  $^{83}\text{Rb}$  was detected by gamma counting in the leachate solutions, however, using a more sensitive analytical technique small amounts of  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  and  $^{241}\text{Am}$  were found in leachates from all soils. It appears that release of this small fraction of mobile radionuclide may have a significant long-term impact on the environment. It also appears that reliance for attenuation of some radionuclides can not be placed solely on characteristics of the soil matrix.

Introduction

Shallow land burial has been used for disposal of low-level radioactive waste material for more than three decades. Recently, investigators have observed small amounts of radioactivity in the environs of several of those sites, signaling the need for a more thorough evaluation of the potential and mechanisms of long-term redistribution of buried radionuclides both on site and off site.

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\*Work performed under the auspices of the Nuclear Regulatory Commission.

present to identify the major clay types as vermiculite and kaolinite. One of the characteristics believed to be very important in mediating the chelate complexation of radionuclides is the available (extractable) iron. Large amounts of extractable iron were present in Fayette and Fergus soils as measured by the synthetic chelating agent diethylenetriaminepentaacetic acid (DTPA).

Leaching columns, 3.8 cm in diameter were prepared in triplicate using 300 g (air dry) of each soil type. A 1-cm plug of washed silica sand (20-50 mesh) was placed at the top and bottom of each column. After packing, each column was saturated with distilled water from the bottom to minimize the trapping of air and then allowed to drain to field capacity prior to commencement of leaching.

Soil column setup and leaching were conducted according to the procedure of Essington and Nishita (Es66). The waste solution was added to the top of the soil column in 10-cm irrigation increments (115 ml each) until the infiltration rate of the waste solution was seriously impaired. At that point 2.5 l of waste solution had been passed through the soil column. Each 10-cm leach was collected and a selected few were analyzed for major gamma emitting radionuclides and  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$ . The gamma emitting radionuclides were measured with a germanium (lithium drifted) detector and pulse height analyzer. The other radionuclides were measured using radiochemical separations and alpha pulse analysis. Electrical conductivity, Eh, pH, and alkalinity were also determined on each leachate. Upon completion of the leaching experiment each column was fractionated into approximately 2-cm increments. Each increment was analyzed for major gamma emitting radionuclides and  $^{238}\text{Pu}$ ,  $^{239,240}\text{Pu}$ , and  $^{241}\text{Am}$ .

The leaching solution was a two-week composite of a liquid radioactive waste material accepted by a waste treatment facility. Table 3 lists several of the many measured characteristics of a similar waste material as collected and after centrifuging to remove particulates greater than about 0.05  $\mu\text{m}$  in diameter. Material remaining in the supernatant solution after centrifugation was designated as "soluble" although the supernatant probably contained small particulates, polymers, and ions. Centrifugation removed substantial quantities of phosphate, aluminum, iron and organic matter, suggesting that a predominance of these materials was associated with particulates larger than 0.05  $\mu\text{m}$ . Those materials are also suspected of being involved in interactions with some of the radionuclides in the waste.

Some of the more prevalent radionuclides present in the waste are listed in Table 4. The ratio of radionuclide concentration before and after centrifuging are given to indicate their degree of partition and to suggest a degree of complexity of the waste solution. Three groupings are suggested in Table 4 based on the amount of radionuclide contained in the soluble fraction relative to the total. The group including  $^{87}\text{Rb}$ , involves the largest amount of soluble radionuclide, whereas the group including  $^{88}\text{Y}$  involves the largest amount of insoluble radionuclide. Note particularly that the partition of the two isotopes of plutonium is significantly different in the two fractions (mean/standard deviation of 4 replicates = C.V. = 0.1).

### Results and Discussion

The distributions of various waste radionuclides in soils and leachates indicate the complexity of the chemical and physical interactions between the

Field experiments to describe the physical and chemical interactions affecting radionuclide migration are very expensive and in some cases cannot yield the information required for prediction. Laboratory tests are, then, generally conducted to derive the necessary data for use in flow or migration prediction models.

Over the past three decades many studies of radionuclide migration have been conducted in soil and geologic materials to assess the degree of sorption or retention the materials have for the radionuclides. However, until recently, most studies of that type relied on single prepared radioisotope solutions in carefully controlled systems and did not represent the very complex conditions existing in radioactive waste interactions with soil or geologic media.

A study was designed to provide information on radionuclide retention and migration in soils as a function of soil type and radionuclide species. Preliminary results show the nature of low-level radioactive waste material interactions with several widely varying soil types. Based on information gained in this study, future experiments will be designed to more precisely evaluate factors affecting retention or migration and to develop a measure of the confidence one might expect in retention values. This information will be incorporated into radionuclide migration and dose assessment models used in low-level waste management and licensing.

This study is one phase of a larger study to derive some confidence in predictions of radionuclide retention by soils and is sponsored by Nuclear Regulatory Commission, Division of Safeguards, Fuel Cycle and Environmental Research. Figure 1 depicts the rationale behind the study. A great deal of variability in the degree of radionuclide retention does exist because of the wide variety of radioactive sources and species in each source and the large variability in the environmental materials available for interaction. The variety of waste sources is indicated in the first column of Fig. 1; examples of characteristics of the waste/soil solution believed to influence retention are listed in the second column. The degree of radionuclide retention as determined by soil types for each waste/soil characteristic will define an envelope from which some confidence relative to retention can be projected. Correlation of the retention with one or a combination of more specific parameters (characteristics), such as, soil texture, pH, clay type, etc., may yield a smaller degree of predicted retention variability. In other words, the more that is known about the system the better one can predict both short- and long-term radionuclide retention.

#### Materials and Methods

Four soils were reacted with a low-level radioactive industrial waste solution in order to evaluate the radionuclide retention capacity of the soils. Several major physical and chemical soil characteristics likely to be related to radionuclide retention are listed in Tables 1 and 2. The A<sub>p</sub> horizon of Fuquay loamy sand represents the top 15 cm of soil collected from the Barnwell area of South Carolina. The A<sub>p</sub> horizon of Fayette silt loam represents the top 18 cm of soil collected from Sheffield, Illinois. The B horizon of Carjo loam and the C horizon of Puye sandy loam are from Los Alamos, New Mexico and were collected from depths of 5 to 15 cm and 25 to 51 cm, respectively. The four soils range in pH (saturated paste) from 5.5 to 7.1 and have similar cation exchange capacities (CEC) ranging from 15 to 23 meq/100 g of soil. Although the clay content of the Fuquay is very low, enough clay is

waste and the soil matrix. That complexity is indicated in Fig. 2 by the presence of at least two forms of  $^{241}\text{Am}$  shown by the accumulation of a large portion in the sand layer and by the small but constant amount in the deeper soil fractions. Figure 2 also shows the  $^{241}\text{Am}$  distribution in the three replicate soil columns of Carjo soil as well as the variability of  $^{241}\text{Am}$  in the various soil fractions. The vertical bars represent the plus or minus one-sigma counting error for each fraction. Note that in the shallow portion of the soil columns the counting error is small and data are reproducible. In the deeper portions of the column the counting error is large, however, the  $^{241}\text{Am}$  levels for the three replicates appears to fall well within the counting error. A non-linear least-squares procedure was used to fit the  $^{241}\text{Am}$  values in the soil column fractions to an exponential equation of the type shown on the figure. Subsequent figures will use those fitted curves where possible for clarity of presentation; however, several of the curves were hand fitted.

Figure 3 shows the distribution of  $^{241}\text{Am}$  in the four soils studied. The distribution of  $^{241}\text{Am}$  in Puye, Carjo, and Fuquay, did not appear to be different, whereas  $^{241}\text{Am}$  in Fayette appeared to be distributed somewhat differently, particularly in the shallower parts of the column. Note that in all cases the sand layer retained about 90% of the  $^{241}\text{Am}$  in the column. Additionally, the level of  $^{241}\text{Am}$  in the deeper column fractions approached a constant. That level of  $^{241}\text{Am}$  was calculated to be nearly the same concentration as that found in the final leachates and therefore, can be attributed to the soil column water remaining at field capacity.

Hafnium-172,  $^{241}\text{Am}$ , and  $^{88}\text{Y}$  appear to be similarly distributed in all soil types as illustrated by the data for the Puye soil in Fig. 4. As with  $^{241}\text{Am}$ , the sand layer accumulated better than 90% of the  $^{172}\text{Hf}$  and  $^{88}\text{Y}$ . Hafnium-172,  $^{88}\text{Y}$ , and  $^{241}\text{Am}$  were not detected in the leachate solutions with the gamma counting technique initially employed. However, the lower levels of  $^{241}\text{Am}$  were easily detected by radiochemical separations and alpha pulse counting.

The distribution of  $^{137}\text{Cs}$  in the four soils is shown in Fig. 5. In this case only about 40% of the  $^{137}\text{Cs}$  was removed by the sand layer; however,  $^{137}\text{Cs}$  was not detected in the leachate solutions. The differential distribution of  $^{137}\text{Cs}$  in the 5- to 12-cm region of the Puye soil may be related to soil characteristics. In the shallower depths the  $^{137}\text{Cs}$  had distributed in a manner similar to that for  $^{241}\text{Am}$ ,  $^{172}\text{Hf}$ , and  $^{88}\text{Y}$  as indicated by the slopes of the distributions in the 0- to 5-cm region.

Strontium-85 distribution in the four soils is shown in Fig. 6. Only about 40% of the  $^{85}\text{Sr}$  was removed by the sand layer and no  $^{85}\text{Sr}$  was detected in the leachate solution. Strontium-85 appears to distribute in the soils differently from the radionuclides discussed earlier. This conclusion is based on the observation that the slopes of the distribution curves in the upper portion of the columns are not as steep as for the other radionuclides, indicating that greater amounts of  $^{85}\text{Sr}$  have moved deeper into the soil columns. Similar distribution patterns were reported by Essington et al. (Es66) and Nishita and Essington (Ni67) in columns of agricultural soils containing radioactive strontium and leached with irrigation or distilled water. Movement or retention of the strontium was believed to be governed by the sorption mechanisms of the soils and the competition of calcium ions for sorption sites. Since no  $^{85}\text{Sr}$  was found in the leachate solutions, that portion of the  $^{85}\text{Sr}$  not removed from the waste leaching solution by filtration appears to be attenuated by sorption mechanisms.

Figure 7 shows the distribution of  $^{83}\text{Rb}$  in the four soils. The distribution of  $^{83}\text{Rb}$  in Puye, Carjo and Fayette was similar to that of  $^{241}\text{Am}$ ; however, only 10% of the  $^{83}\text{Rb}$  was retained by the sand layer and no  $^{83}\text{Rb}$  was found in the leachates.

Rubidium-83 distribution in Fuquay was quite different from that in the other soils in that concentrations were constant with depth to about 7 cm and then steadily decreased to a non-detectable level. It appears that the  $^{83}\text{Rb}$  was retained by Fuquay to a lesser degree than was  $^{85}\text{Sr}$  (Fig. 6). However, based on the similarity of the  $^{83}\text{Rb}$  distribution to that of  $^{85}\text{Sr}$  similar sorption mechanisms may be involved. No effort was made to determine stable rubidium in the waste solution; stable rubidium could have a significant effect on  $^{83}\text{Rb}$  retention by the soil matrix.

Plutonium-238 and  $^{239,240}\text{Pu}$  were measured in the soil column leachates only. Figure 8 shows the amount of  $^{238}\text{Pu}$  added with each increment of waste solution and the amount of  $^{238}\text{Pu}$  found in each leachate; data are presented as averages of the leachates from the three soil columns. The appearance of  $^{238}\text{Pu}$  in the first leach indicates a rapid breakthrough. The attainment of a relatively constant level after the second waste addition indicates that portion of  $^{238}\text{Pu}$  in the waste solution that was not filtered or sorbed by the soil. A comparison of the  $^{238}\text{Pu}$  in the leachate with that added to the soil columns shows that 83 to 97% of the  $^{238}\text{Pu}$  was retained by the soil columns. Those results indicate that there was a small but highly mobile fraction of  $^{238}\text{Pu}$  in the waste solution. Although  $^{239,240}\text{Pu}$  was found in the same leachate,  $^{239,240}\text{Pu}$  reacted somewhat differently from  $^{238}\text{Pu}$ . This is shown in the ratios of  $^{239,240}\text{Pu}$  to  $^{238}\text{Pu}$  in the waste solution, centrifuged waste, and the soil leachates (Fig. 9). In general, the ratios for soil leachates fell between those of the source waste and centrifuged waste solutions. In the early stages of leaching there were rapid changes in the leachate plutonium ratios, but as leaching progressed the ratios tended to converge at about 0.1. Those data show that isotopes of the same element can react differently in the soils. Plutonium in the waste solution originates from a number of widely different operations. Those operations dealing primarily with  $^{238}\text{Pu}$  tend to dispose of a higher percentage of soluble  $^{238}\text{Pu}$  than soluble  $^{239,240}\text{Pu}$ . Those operations dealing primarily with  $^{239,240}\text{Pu}$  and  $^{241}\text{Am}$  (Table 4) tend to dispose of particulate material or material prone to be associated with particulates. Thus the nature of the source material may dominate the retention pattern in soils and the differences in plutonium isotopic distributions are not necessarily due to basic difference in behavior of the isotopes.

The waste/soil system is complex as indicated by the data presented. That complexity may be a result of a number of interactive mechanisms. The major mechanisms, in addition to filtration and ion exchange, include: precipitation and dissolution of calcium carbonate; complexation; and microbiological growth. Those mechanisms as well as evaluation of changes in the waste source upon aging and results of batch sorption studies with the same soils were reported by Fowler *et al.* (Fo78).

Waste radionuclides interacting with calcium carbonate, the primary inorganic mineral in the waste, should behave in a manner similar to that of the calcium carbonate. For example, changes in the carbonate system which result in either a precipitation or dissolution of solid carbonates should result in

a change in the radionuclide concentration in the soil solution. Such changes have been noted to occur in the waste solution upon aging, and undoubtedly occur upon contact with the soil. The changes could continue to occur during the progress of leaching as the leaching solution aerates or changes upon encountering different materials in the soil column. The extent of the carbonate effect on radionuclide retention was not specifically investigated in the experiment. However, the presence of  $\text{CaCO}_3$  in the waste solutions was confirmed by X-ray diffraction. Significant decreases in the concentration of soluble plutonium and americium were observed upon formation of  $\text{CaCO}_3$  in the waste material, whereas, changes in the soluble cesium and uranium concentrations did not occur. Those observations may account, in part, for the differences in the amounts of  $^{241}\text{Am}$  and  $^{137}\text{Cs}$  retained by the sand layer on top of the soil column.

The small but significant amount of plutonium and americium found in the leachate solutions may be due to carbonate complexing or the presence of chelated species. The charge of some soluble actinides in the waste was shown to be predominately negative whereas the charge of the  $^{137}\text{Cs}$  was predominantly positive (Po79). This was accomplished by passing the soluble waste fraction through cation and anion exchange resins and observing the amount of radionuclide not held by the resin (Fowler *et al.* Fo78). The work of Alberts *et al.* (Al77) on the identification of the charge of plutonium in samples of Lake Michigan water indicated that the charge of the plutonium species was negative. They attributed the negative nature of the charge to the formation of a stable carbonate complex in the  $\text{CaCO}_3$  super-saturated water having a pH of approximately 8. Ames *et al.* (Am76) present stability diagrams for plutonium and uranium which indicate that at pH 8.0 carbonate complexes could account for the predominantly negative charge of plutonium and uranium species.

Although the waste solution was not analyzed for the presence of chelating agents it is strongly suspected that significant quantities were present. The sources of the waste solution include decontamination fluids containing detergents and chelating agents and chemical laboratory wastes where chelating agents and other organic complexers are used and discarded. The effect of the chelating agents would be to form a very stable complex with the radioactive rare earth or actinide ions present and at pH 8 the resulting complexes are likely to be neutral or negatively charged. Those species would migrate through the soil rapidly with the possibility of exchange with metal ions in the soil solution. The degree of exchange of the radioactive ion with a metal ion would be dependent upon their concentrations in the soil solution and upon their relative stabilities with the chelate ligand.

During the conduct of the column leaching experiment an algal growth was allowed to be established in the columns. Normally the leaching experiment would be conducted with care not to allow unnatural microbiological activity; however, waste solutions allowed to impact the open environment would surely be subject to microbiological activity. The effect of the algae on radionuclide retention in the soil columns is a valid parameter. A separate experiment was conducted to test the effect of algal bloom was allowed in the waste solution in which the pH was adjusted to 6.0; the algae were removed, washed, and analyzed. The effect of algae on retention of waste radionuclides depended on the radionuclide; a large percent of the actinides (86-95%), but only 30% of the cesium was associated with the algae. Those results are consistent with the large



accumulation of  $^{241}\text{Am}$ ,  $^{172}\text{Hf}$ , and  $^{88}\text{Y}$  and the somewhat smaller degree of accumulation of  $^{137}\text{Cs}$ ,  $^{85}\text{Sr}$ , and  $^{83}\text{Rb}$  in the sand layer on the soil column.

The information presented thus far is indicative of the complexity of the waste/soil systems. Reactions of liquid waste materials from different low level waste streams, shallow waste burial pits or trenches, or the high level waste processing streams accidentally released to the soil environment may be even more complex.

Relating the retention data to migration rates and distance is generally accomplished by use of the distribution coefficient ( $K_d$ ) in various velocity models. A simple version of a model relating radionuclide velocity ( $V_R$ ) to  $K_d$  is as follows:

$$V_R = \frac{V_W}{1 + K_d r}$$

where:  $r$  = bulk density/porosity,  
 $V_W$  = velocity of water.

More complex models have been derived, some of which are reviewed by Travis (Tr78). Most velocity models still use  $K_d$  as the parameter that transmits the degree of radionuclide retention to the model. Actually  $K_d$  was defined by Mayer and Thompkin (Ma47) and is as follows:

$$K_d = \left( \frac{C_0}{C} - 1 \right) \frac{V}{M}$$

where:  $C_0$  = tracer concentration in solution before adding mineral,  
 $C$  = tracer concentration in liquid phase of mineral-water suspension,  
 $V$  = volume of liquid,  
 $M$  = mass of solid.

Many investigators recognize that the  $K_d$  values include a number of assumptions or qualifications which result in minimal impact when considering single source radionuclides in simple systems. The inherent assumptions and qualifications appear to not result in minimal impact when used for real radioactive wastes. Quoted  $K_d$  values might be used by licensers or policy makers without due consideration of the limitations.

The distribution coefficient should be independent of the solid to liquid ratio. In practice, however, equilibrium is never attained in an environmental system, and evidence has appeared suggesting that  $K_d$  is not independent of the solid to liquid ratio. This has profound impact when measuring  $K_d$  in the laboratory using a 4-part liquid to 1-part solid ratio then applying that data to a system of water flow in a similar soil or geologic medium where the effective water to solid ratio may be 1 to 100. Further, a more important concern in the concept of  $K_d$  for predicting radionuclide migration relates to the complexity of the waste/soil system. A

number of different forms of a given radionuclide were shown to exist e.g., filterable, sorbable, and highly mobile. Each form may consist of a number of species; each specie could be described by a separate  $K_d$  value. Using  $K_d$  values generated on a combined system will not predict the degree of retention of all of the species present. Some species will migrate more rapidly and some more slowly than average. Those species migrating rapidly, even if in low concentrations, may constitute a significant impact on the environment. Even if a  $K_d$  value is generated for a particular specie, as that specie migrates to a new matrix material or is intercepted by a leach solution of different characteristics, the degree of retention will likely change thus negating that  $K_d$  as a predictor.

### Summary

Four soils of widely differing characteristics were treated with a radioactive liquid waste solution in order to evaluate the degree and pattern of retention of various long lived radionuclides. The experimental system was designed to provide preliminary information on various waste/soil systems representative of accidental releases to the environment and possible releases from shallow land burial.

Results of the studies indicated that the wastes were very complex in form and species of each radionuclide. Some isotopes of the same element appear in different forms, each acting differently with respect to retention by soils. The results also suggest that extreme care must be exercised when generating and using the standard distribution coefficient ( $K_d$ ) in migration and dose assessment models. The  $K_d$  values generated in laboratory tests may not predict adequately the true degree of radionuclide interaction with the soil. Migration rates of some portion of the radionuclide sources may change with time and distance complicating even further the predictive value of  $K_d$ .

Results of the laboratory studies are significant in the interpretation of the migration of waste radionuclides from shallow land burial pits. Although wastes buried in a shallow land disposal pit are considered solids, a certain fraction is converted to a liquid phase through decomposition processes and the action of seepage water and becomes available for transport. Physical removal of a filterable fraction by a surface soil in a biochemically dynamic environment will result in eventual degradation of organic matter and the release of associated radionuclides to the soil solution.

The fate of all forms of a radionuclide in the environment may ultimately be similar. However, initially some radionuclides may migrate great distances because of their nature. Thus, a prediction of the movement or changes in the movement of waste radionuclides from a point of release necessitates some knowledge of biochemical, chemical, and physical interactions of waste radionuclides with their environment.

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Fig. 1. Flow diagram relating radioactive waste source and soil characteristics to radionuclide retention and the development of statistically based correlations.

Fig. 2. Distribution of  $^{241}\text{Am}$  in columns of Carjo soil after leaching with radioactive waste.  $A/A_0$  represents fraction of total column  $^{241}\text{Am}$  found in each increment.

Fig. 3. Distribution of  $^{241}\text{Am}$  in four soils.

Fig. 4. Comparison of the distribution of  $^{241}\text{Am}$ ,  $^{172}\text{Hf}$  and  $^{88}\text{Y}$  in Puye soil.

Fig. 5. Distribution of  $^{137}\text{Cs}$  in four soils.

Fig. 6. Distribution of  $^{85}\text{Sr}$  in four soils.

Fig. 7. Distribution of  $^{83}\text{Rb}$  in four soils.

Fig. 8. Plutonium-238 in source waste and in leachates from four soils.

Fig. 9. Ratio of  $^{239},^{240}\text{Pu}$  to  $^{238}\text{Pu}$  in source waste, in centrifuged waste, and in leachates from four soils.

Table 1. Selected major physical characteristics of four soils used in leaching studies.

Table 2. Selected major chemical characteristics of four soils used in leaching studies.

Table 3. Selected physical and chemical properties of source waste (TOTAL) and solution phase (SOLUBLE) after centrifugation.

Table 4. Major radionuclides identified in source waste, the radioactive decay half-life,  $T_{1/2}$ , and the partition upon centrifugation.

Fig. 1. Flow diagram relating radioactive waste source and soil characteristics to radionuclide retention and the development of statistically based correlations.

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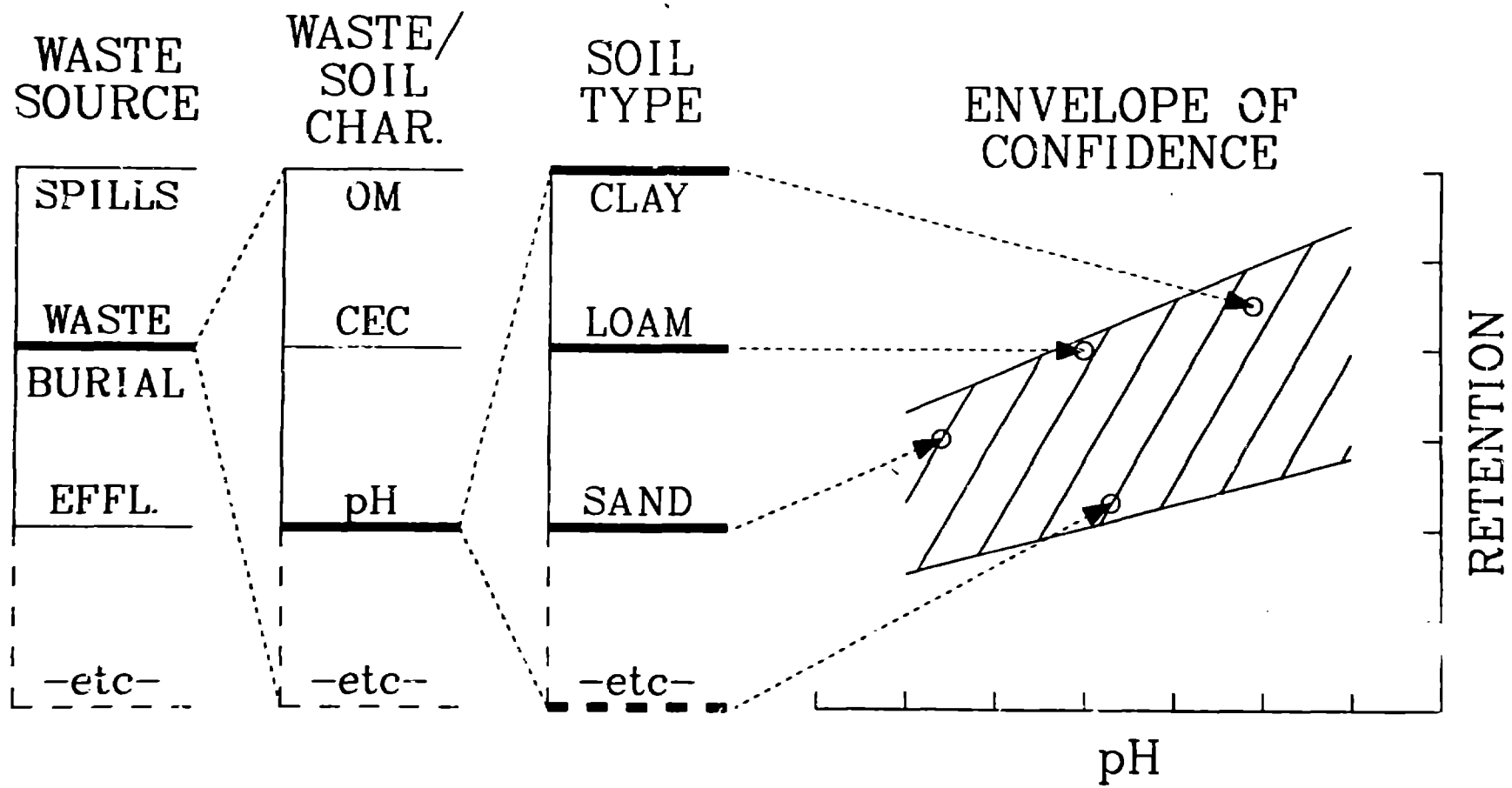


Table 1. Selected major physical characteristics of four soils used in leaching studies.

	Puye	Carjo	Fayette	Fuquay
$P_w$ at F.C. <sup>1</sup>	43	40	40	30
CEC	23	18	15	16
pH	7.1	6.4	6.6	5.5
SAND %	45	53	19	86
SILT %	48	37	73	14
CLAY %	7.5	10	8.5	~ 0
CLAY TYPE <sup>2</sup>	KM	KM	KMV	VK

<sup>1</sup>Percent Moisture at Field Capacity

<sup>2</sup>V=Vermiculite; K=Kaolinite; M=Montmorillonite



Table 2. Selected major chemical characteristics of four soils used in leaching studies.

	Puye	Carjo	Fayette	Fuquay
	(ppm)	(ppm)	(ppm)	(ppm)
Total Ca	10 000	10 000	10 000	1000
Ammonium				
Acetate				
Extractable				
Ca	1800	1000	1400	230
Mg	170	200	330	22
Fe	4.8	5.4	36	1
Mn	24	54	24	16
Al	6	9	24	4
DPTA Extr. Fe	6	7	28	50

Table 3. Selected physical and chemical properties of source waste (TOTAL) and solution phase (SOLUBLE) after centrifugation.

	TOTAL	SOLUBLE
pH	7.8	8.1
Eh (mV)	+410	+400
PO <sub>4</sub> (*)	29	2
Ca (*)	75	51
Al (*)	2	0.2
Fe (*)	12	0.4
COD (*)	170	60

(\*)=(ppm)

Table 4. Major radionuclides identified in source waste, the radioactive decay half-life,  $T_{1/2}$ , and the partition upon centrifugation.

	$T_{1/2}$	$S/T^1$
$^{83}\text{Rb}$	83d	.67
$^{85}\text{Sr}$	64d	.54
$^{137}\text{Cs}$	30y	.60
$^{238}\text{U}$	$4.5 \times 10^9 \text{y}$	.46
$^{88}\text{Zr}$	85d	.15
$^{60}\text{Co}$	5.26y	.11
$^{88}\text{Y}$	108d	.018
$^{172}\text{Hf}$	5y	.001
$^{241}\text{Am}$	458y	.012
$^{238}\text{Pu}$	86.4y	.031
$^{239,240}\text{Pu}$	24390y, 6580y	.011

$^1T$ =Source waste, S=Soluble fraction

Fig. 2. Distribution of  $^{241}\text{Am}$  in columns of Carjo soil after leaching with radioactive waste.  $A/A_0$  represents fraction of total column  $^{241}\text{Am}$  found in each increment.

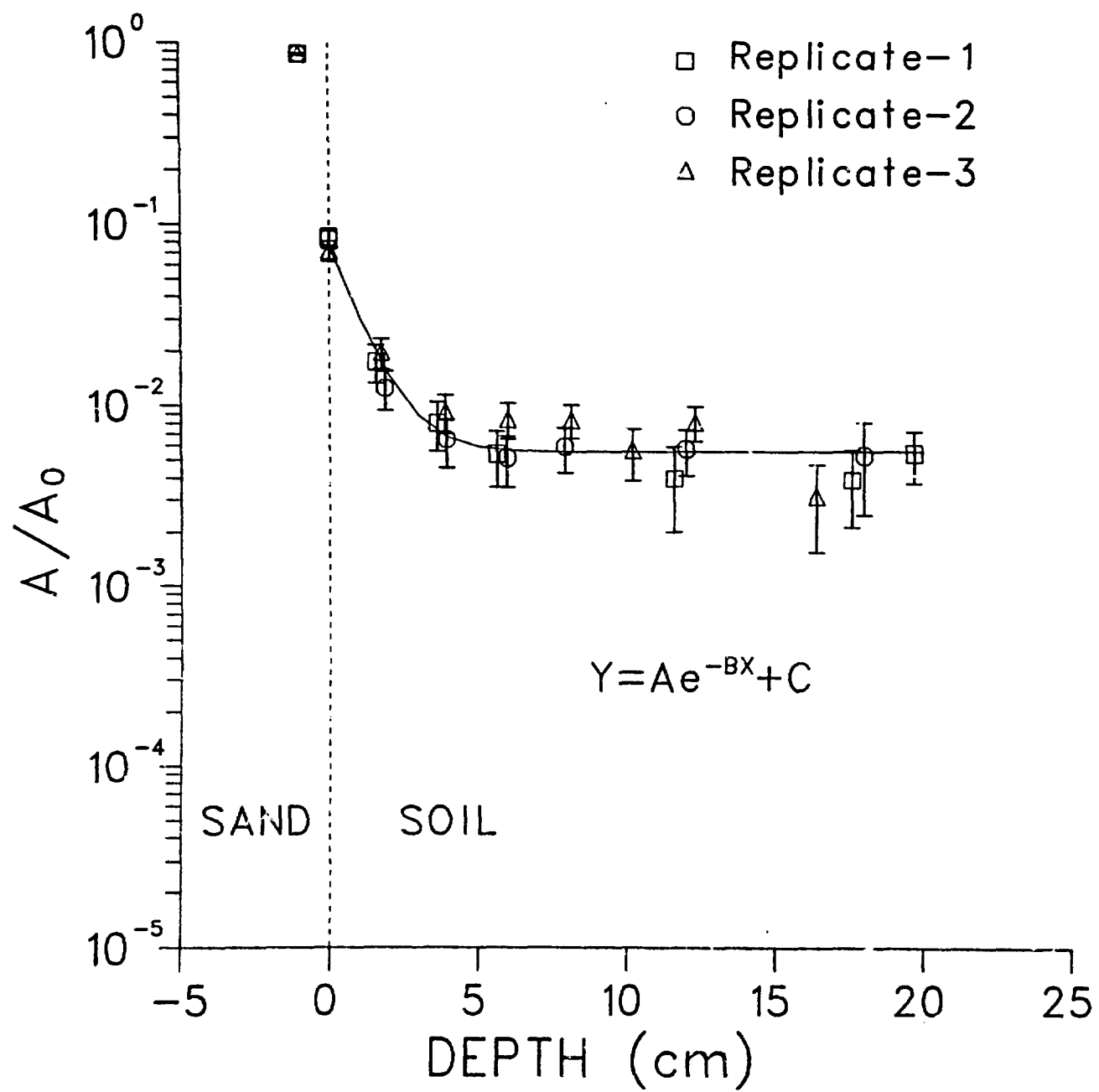




Fig. 3. Distribution of  $^{241}\text{Am}$  in four soils.

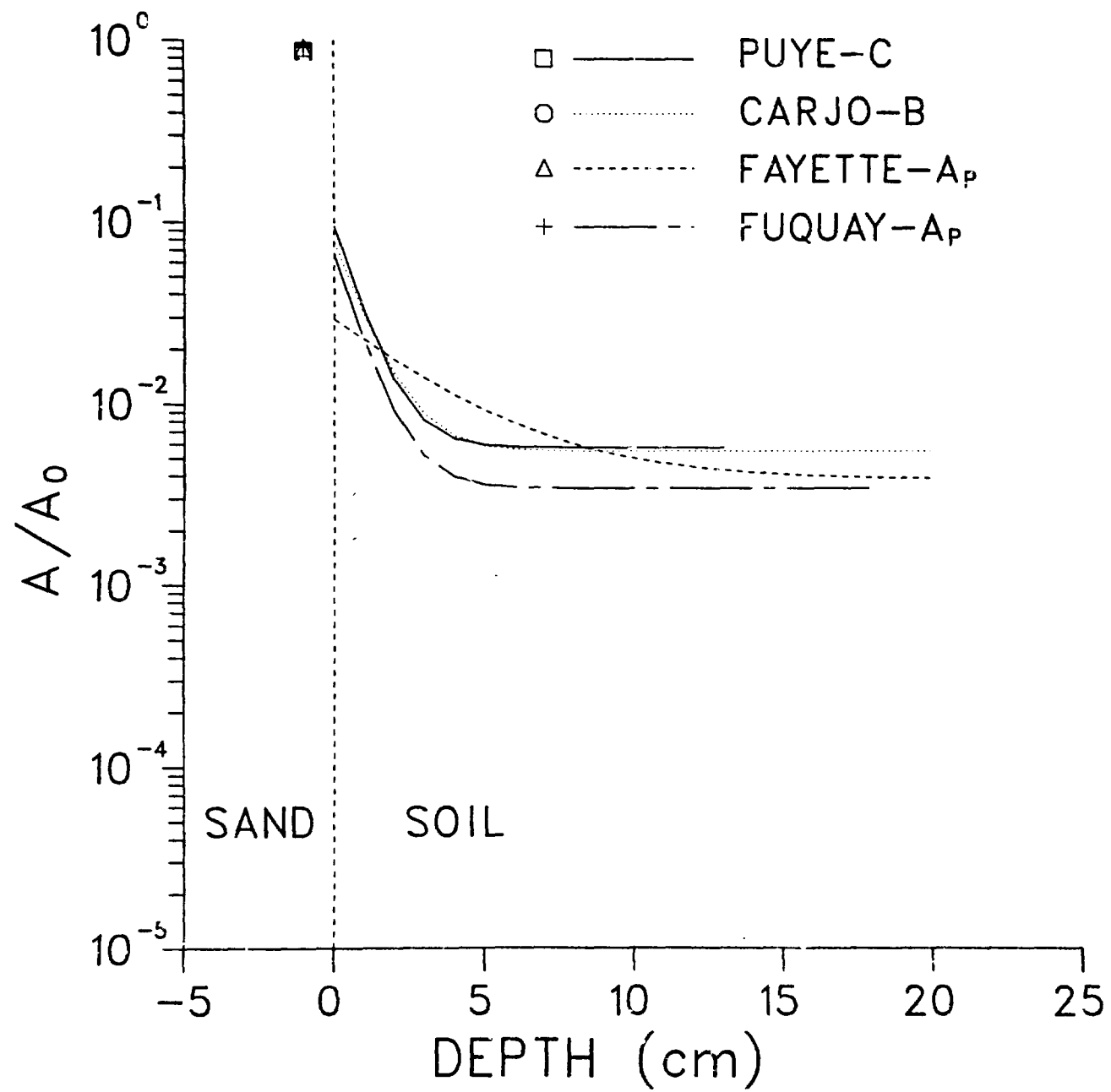


Fig. 4. Comparison of the distribution of  $^{241}\text{Am}$ ,  $^{172}\text{Hf}$  and  $^{81}\text{Y}$  in Puyé soil.

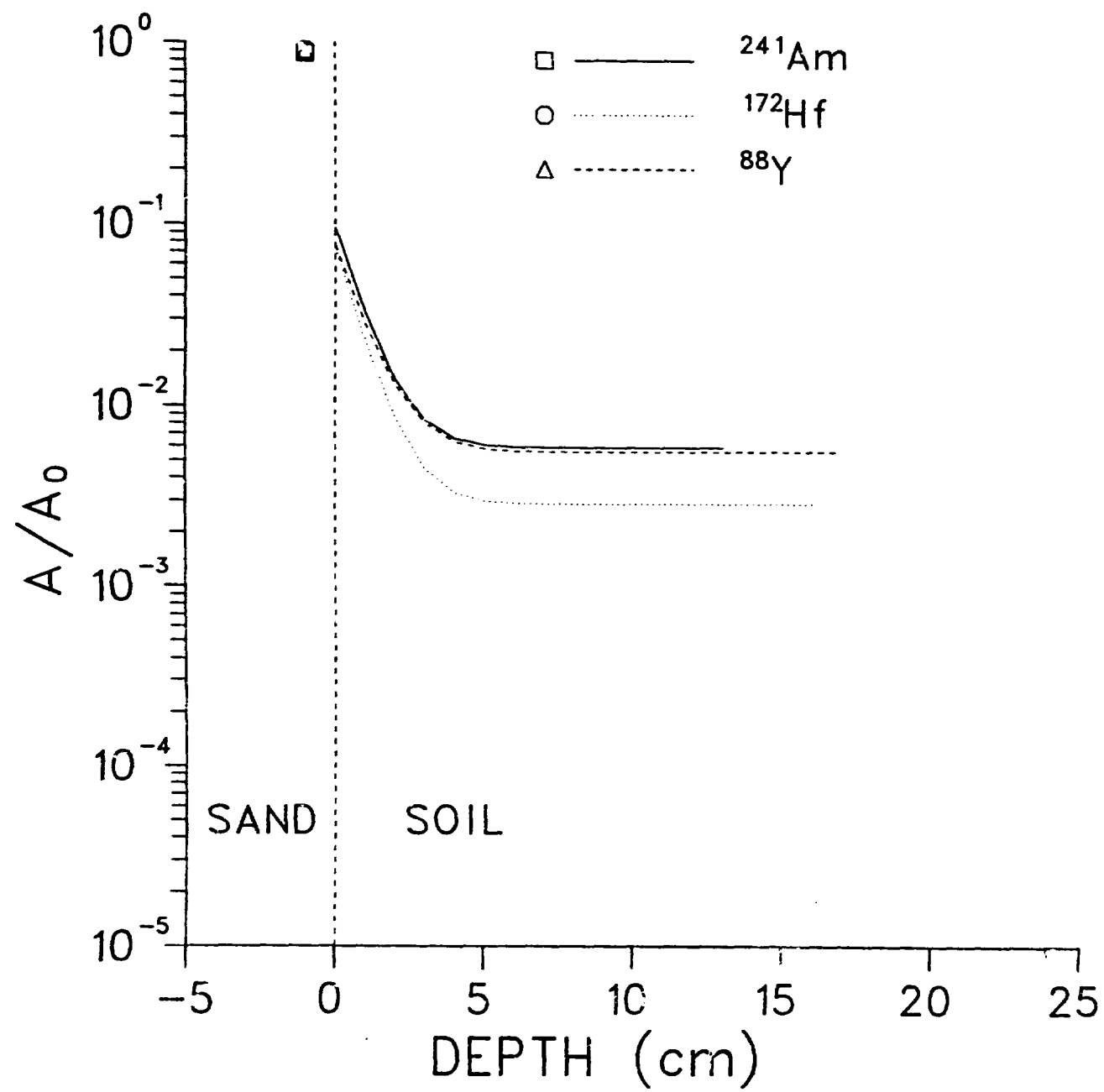


Fig. 5. Distribution of  $^{137}\text{Cs}$  in four soils.

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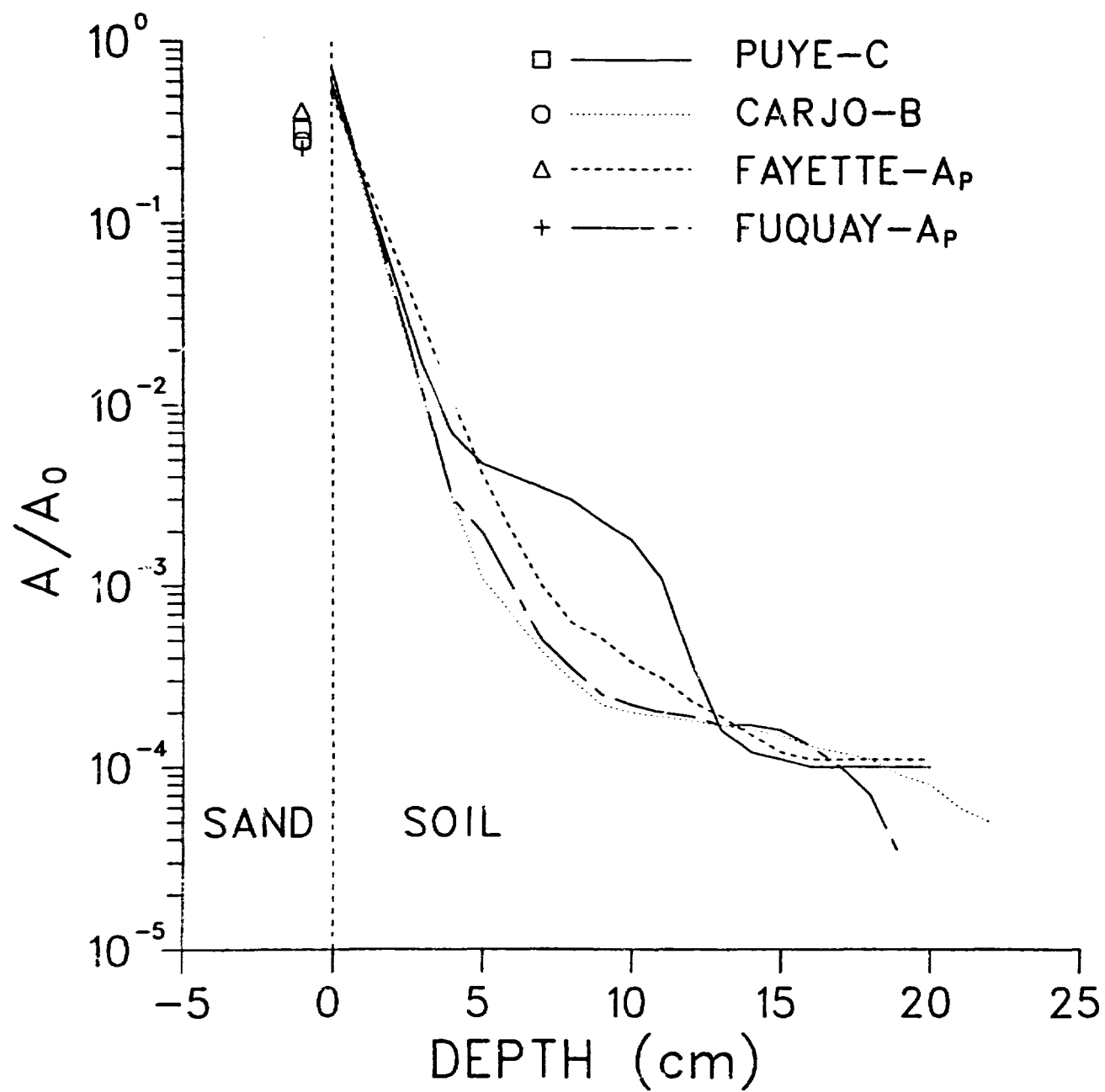


Fig. 6. Distribution of  $^{85}\text{Sr}$  in four soils.

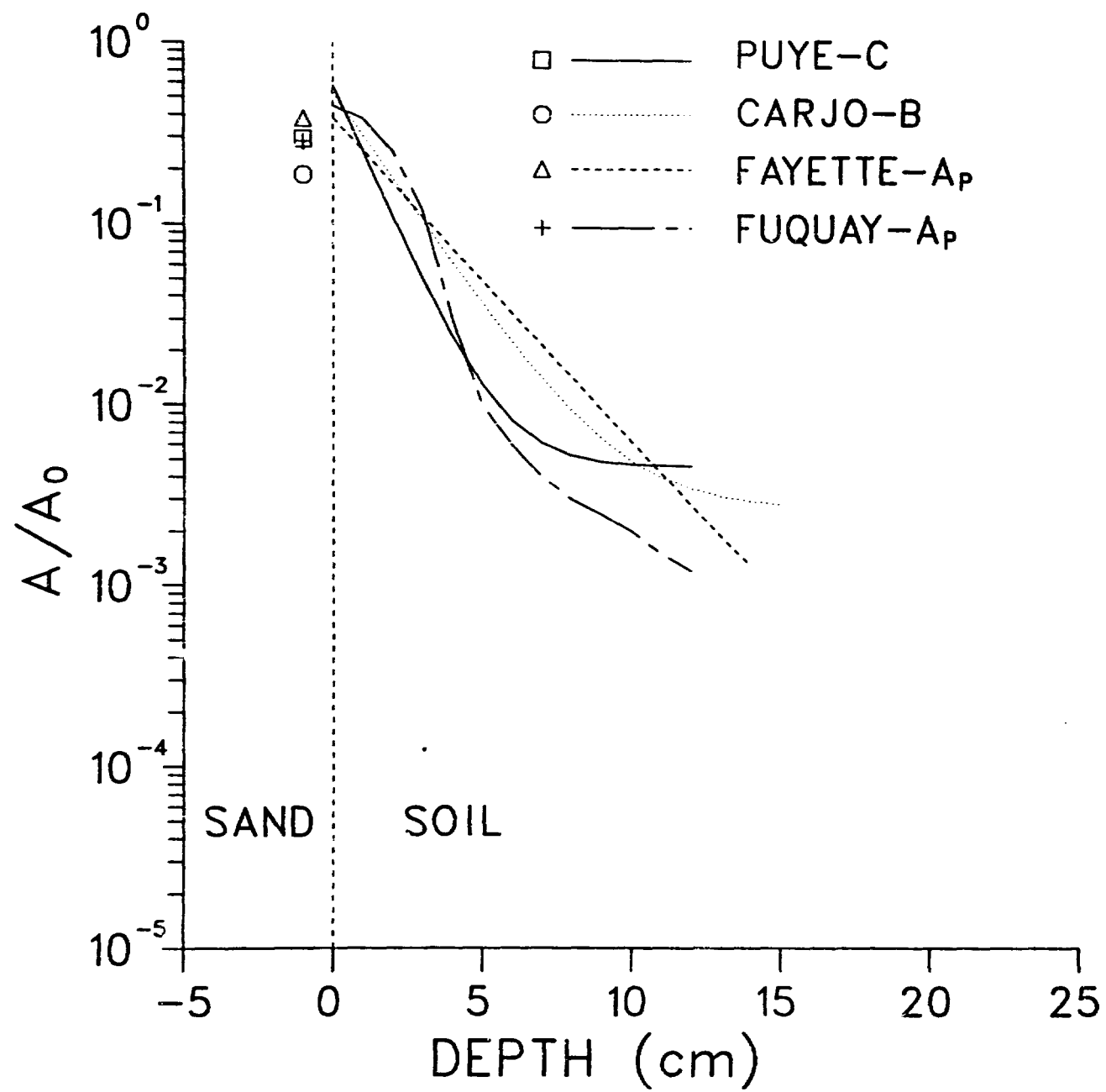




Fig. 7. Distribution of  $^{83}\text{Rb}$  in four soils.

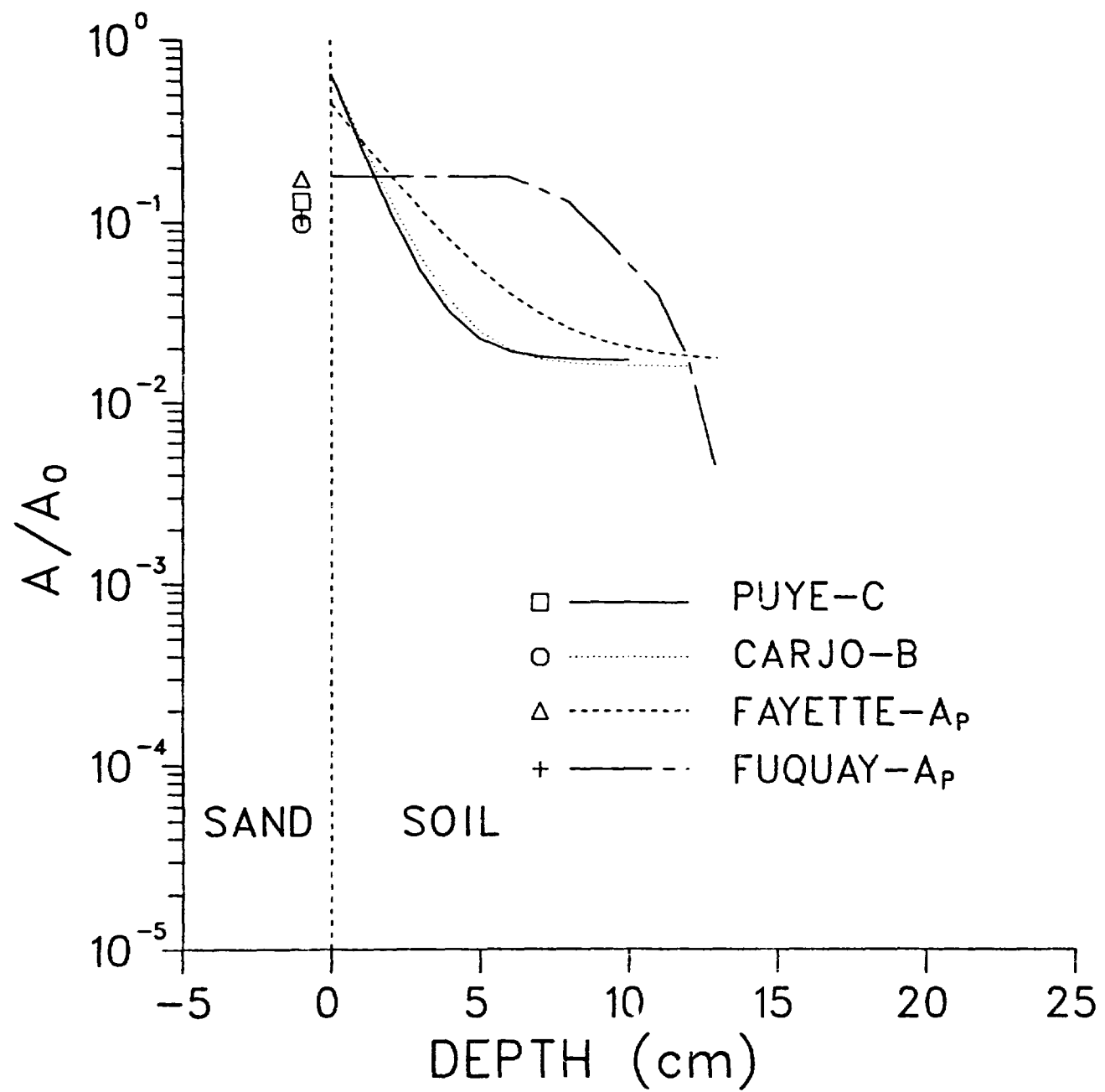


Fig. 8. Plutonium-238 in source waste and in leachates from four soils.

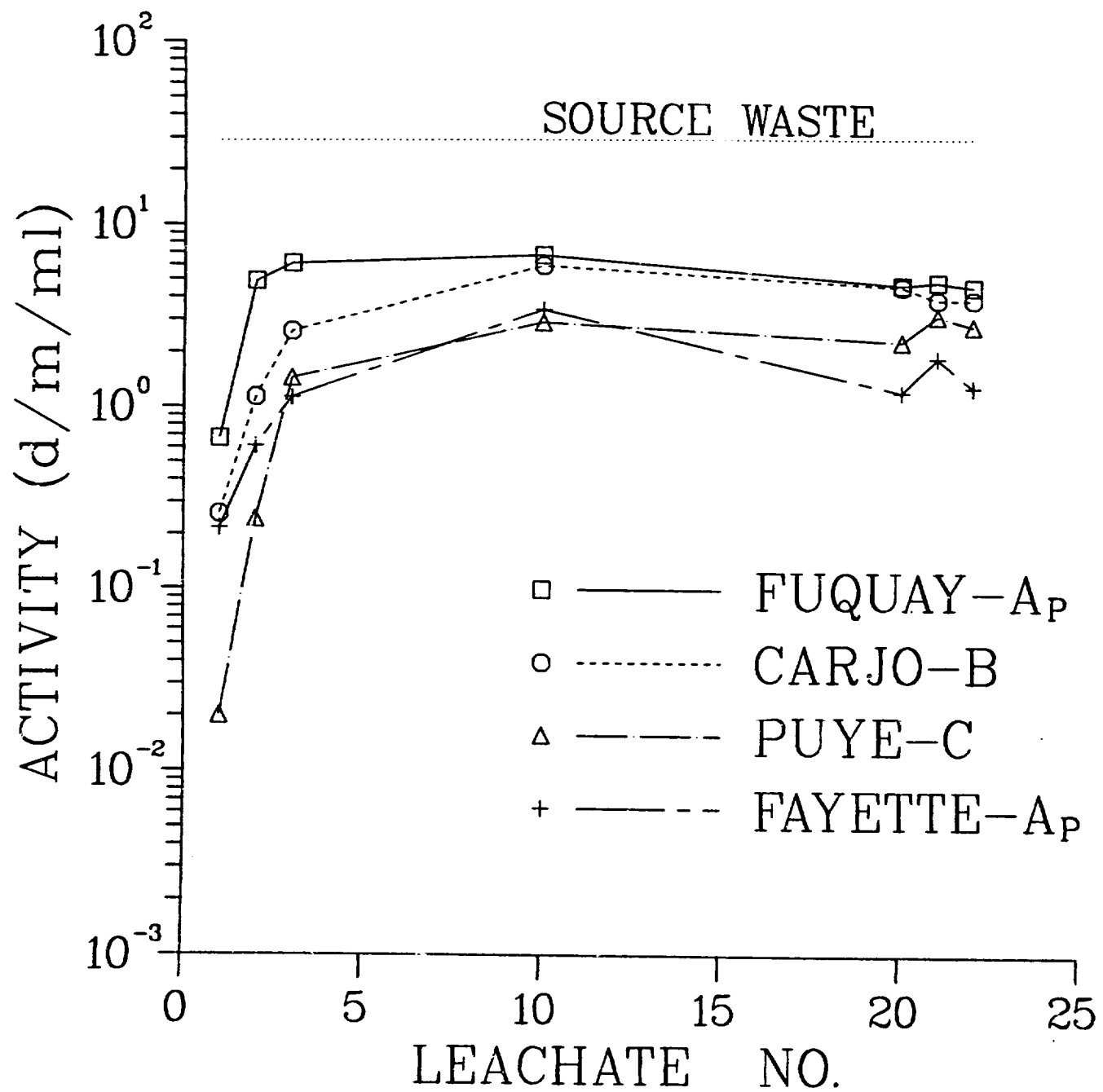


Fig. 9. Ratio of  $^{239,240}\text{Pu}$  to  $^{238}\text{Pu}$  in source waste, in centrifuged waste, and in leachates from four soils.

